



#12  
3/12/96  
PATENT

IN THE UNITED STATES PATENT  
AND TRADEMARK OFFICE

Applicants:	)	For: Cementitious Gypsum-
	)	Containing Compositions
Stav et al.	)	and Materials Made
	)	Therefrom
Serial No.: 08/384,756	)	
	)	
	)	Group Art Unit: 1108
Filed: February 7, 1995	)	Examiner: M. Marcheschi

DECLARATION OF ELISHA STAV

ELISHA STAV hereby states as follows:

1. I am a named co-inventor of the above-identified application. I received a PhD in Chemistry and Building Materials from the Israel Institute of Technology in 1993. I also have received an MS in Chemistry and Polymer Science and a BS in Chemistry. I am currently employed by National Gypsum Company, the assignee of the patent application. My current title is Senior Chemist. I have been involved in process research concerning gypsum-containing and/or cementitious compositions for National Gypsum for about 2 1/2 years.

2. I have read and am familiar with the official action of September 8, 1995, as well as the references applied therein against the claims of the application.

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GROUP 110

3. With respect to the Bentur et al. manuscript entitled "Gypsum of Improved Performance Using Blends With Portland Cement and Silica Fume" applied by the examiner in the September 8, 1995 official action, I received the manuscript from Dr. Bentur on July 12, 1993. At that time, I did not know whether or not the manuscript had been published.

4. In connection with the development of the invention claimed in the application, I directly supervised a laboratory testing procedure to compare compositions according to the invention recited in the claims with similar compositions wherein the pozzolanic aggregate (i.e., filler) component of the composition was replaced by a non-pozzolanic aggregate of similar size (TEST I) and where a composition according to the invention recited in the claims was compare to a similar composition which did not include silica fume (TEST II).

#### TEST I

5. The following "Table A" discloses two "master blends," hereafter referred to as MB 1 and MB 2, each including gypsum (stucco), Portland cement, and silica fume which were prepared and then blended with either pozzolanic aggregate according to the invention or a calcium carbonate (non-pozzolanic) aggregate.

TABLE A

MASTER BLEND	Gypsum (Stucco) (wt.%) <sup>1</sup>	Portland Cement (wt.%) <sup>2</sup>	Silica Fume (wt.%) <sup>3</sup>
MB 1	40	46	14
MB 2	74	20	6

1. Calcidine Stucco (Savannah, Georgia)
2. Portland Cement Type-3
3. Silica Fume (SKW, Niagara Falls, NY)

6. About 75 wt.% of each of MB 1 and MB 2 were mixed with about 25 wt.% pumice. Also, about 75 wt.% of both MB 1 and MB 2 were each mixed with about 25 wt.% calcium carbonate. The calcium carbonate utilized had approximately the same particle size as the pumice aggregate. Each of the four resulting mixtures was mixed with about 60 wt.% water. All four compositions were allowed to dry at about 70 degrees F and a relative humidity of about 50%. The compressive strengths of samples (2 inch by 2 inch by 2 inch cubes) of each of the compositions were tested after 1, 3, 7, and 28 days by pressing in an Instron press according to ASTM C472-9A. The results of these tests are shown in Table B.

TABLE B

Aggregate	Pumice <sup>1</sup>			Calcium Carbonate (Pumice Size) <sup>2</sup>		
	Density (Lbs/cu.ft)		Compressive Strength (PSI)	Density (Lbs/cu.ft)		Compressive Strength (PSI)
Curing Days	Wet <sup>3</sup>	Dry	Wet	Wet	Dry	Wet
1	102.3	79.8	1151	110.3	87	725
3	102.2	83.3	1779	109.5	88.9	1329
7	102.2	83.3	2646	110.7	92.6	2155
28	102.1	84.8	4267	110.1	92.8	3983
MB 2						
Curing Days						
1	101.4	78.4	668	108.5	85.4	788
3	101.5	79.6	975	108.9	85.8	913
7	101.2	79.5	1254	107.7	84.8	1134
28	101.6	79.8	2221	108.5	84.8	1979

1. 3H Pumice from Hess Pumice Products, Inc.  
 Exhibiting the following passing amounts per mesh size:  
 Mesh size: 14 30 40 50 60  
 Pass (%) 100 11-31 0-19 0-9 0-7

2. Carbonate Z from Georgia Marble.  
 Maximum % retained: 2 on 10 mesh.  
 Maximum % passing: 10 through 20 mesh.

3. Wrapped in plastic during cure.

7. About 75 wt.% of each of MB 1 and MB 2 also were mixed with about 25 wt.% fillite. Also, about 75 wt.% of both MB 1 and MB 2 were each mixed with about 25 wt.% calcium carbonate which had approximately the same particle size as the fillite aggregate. Each of the four resulting mixtures was mixed with about 60 wt.% water. All four compositions were allowed to dry at about 70 degrees F and a relative humidity of about 50%. The compressive strengths of samples (2 inch by 2 inch by 2 inch cubes) of each of the compositions were tested after 1, 3, 7, and 28 days by pressing in an Instron press according to ASTM C472-9A. The results of these tests are shown in Table C.

TABLE C

Filler	Filllite <sup>1</sup>			Calcium Carbonate (Filllite Size) <sup>2</sup>		
MB 1	Density (Lbs/cu.ft)		Compressive Strength (PSI)	Density (Lbs/cu.ft)		Compressive Strength (PSI)
	Wet <sup>3</sup>	Dry	Wet	Wet	Dry	Wet
1	79.7	62.2	822	110.6	86.4	759
3	78.6	63.4	1279	108.7	86	1554
7	78.7	64.7	2313	106.8	90	2492
28	78.8	65.9	4154	108.9	93	4196
MB 2						
Curing Days						
1	78.2	60.3	492	108.1	83.8	838
3	77.3	60.4	780	108.8	85	988
7	78.2	61.2	1063	107.1	84.5	1100
28	77.6	61.7	2451	108.7	86.4	2163

1. Filllite 500 from Filllite Division of Boliden Intertrade, Inc., Atlanta, Georgia.  
Hollow silicate spheres with particle size ranging between 5 and 300 microns.
2. Marb'Blend from Georgia Marble.  
Maximum % retained: 1 on 40 mesh.  
Maximum % passing: 42 through 200 mesh.
3. Wrapped in plastic during cure.

8. As illustrated in Figs. I and II attached to this paper, at the end of 28 curing days, compositions according to the invention exhibited greater compressive strengths than similar compositions utilizing similarly sized non-pozzolanic aggregate.

9. Moreover, as illustrated in Figs. III and IV attached to this paper, each of the compositions according to the invention had a density considerably less than the density of the calcium carbonate-filled counterpart composition. Yet, after 28 curing days, each of the compositions according to the invention exhibited a compressive strength greater than its calcium carbonate-filled counterpart.

10. Scanning electron microscope (SEM) micrographs also were taken of the samples. The photographs attached to this paper are as follows:

Photo 1: 100x magnification of an MB 1 and 3H pumice composition according to the invention described in Tables A and B and paragraph 7, *supra*.

Photo 2: 200x magnification of composition shown in Photo 1.

Photo 3: 500x magnification of the composition shown in Photo 1.

Photo 4: 5,000x magnification of an MB 1 and Fillite 500 composition according to the invention described in Tables A and C and paragraph 8, *supra*.

Photo 5: same as Photo 4.

Photo 6: 15,000x magnification of the composition shown in Photo 4.

Photo 7: another 15,000x magnification of the compositions shown in Photo 4.

Photo 8: 500x magnification of an MB 1 and calcium carbonate composition described in Tables A and C and paragraph 8, *supra*.

11. With respect to Photos 1, 2, and 3, there does not appear to be any separation between the pumice and the surrounding matrix.

12. With respect to Photos 4-7, there also does not appear to be separation between the fillite particles and the surrounding matrix. In particular, there appears to be a halo effect 100 between the fillite particle 200 and the surrounding cementitious matrix 300.

13. An EDXRA analysis confirmed the presence of silicon at the fillite/cementitious matrix interface (i.e., the halo area 100 shown in the photos), suggesting that this CSH are silicon rich, probably from the silica fume. The



EDXRA analysis is attached as Figs. V to VII and illustrates the following:

Fig. V: EDXRA Chart of a Fillite Particle (element 200 in photos) shows SI wt.% of 30.62.

Fig. VI: EDXRA Chart of a Fillite Particle / Cementitious Matrix Interface (halo area 100 in photos) shows SI wt.% raised to 53.67.

Fig. VII: EDXRA Chart of a Cementitious Matrix located adjacent to a Fillite Particle/ Matrix Interface (area 300 in photos) shows SI wt.% down to 35.47.

14. In contrast, with respect to Photo 8, when calcium carbonate is used in a composition instead of a pozzolanic aggregate, the calcium carbonate does not appear to interact with the surrounding matrix and separations are evident throughout the sample.

## TEST II

15. A composition  $\alpha$  according to the claims of the application was prepared with the components set forth in the amounts stated in Table D below. A cementitious composition  $\beta$  with components set forth in the amounts stated in Table D below (which did not include silica fume) also was prepared.

TABLE D

Material	Composition $\alpha$	Composition $\beta$
Beta-Gypsum (Stucco)	60	60
Type III Portland Cement	20	20
Fillite 500 Pozzolan Aggregate <sup>1</sup>	15	20
Silica Fume	5	0

1. Fillite Division of Boliden Intertrade, Inc., Atlanta Georgia; typical particle size of 5-300 microns.

16. The compositions  $\alpha$  and  $\beta$  were each mixed with about 60 wt.% water. Both compositions were allowed to dry at about 70 degrees F and a relative humidity of about 50%. The compressive strengths of samples (2 inch by 2 inch by 2 inch cubes) of each of the compositions were tested after 28 days by pressing in an Instron press according to ASTM C472-9A.

17. The results of the tests and the densities of each of the compositions are shown in Table E. The composition  $\alpha$  according to the claims of the invention exhibited a greater compressive strength than the otherwise similar composition  $\beta$  which did not include silica fume.

TABLE E

Composition	$\alpha$	$\beta$
Compressive Strength (dry) (PSI)	3150	2500
Density (dry) (Lbs/cu.ft.)	65.7	60.1

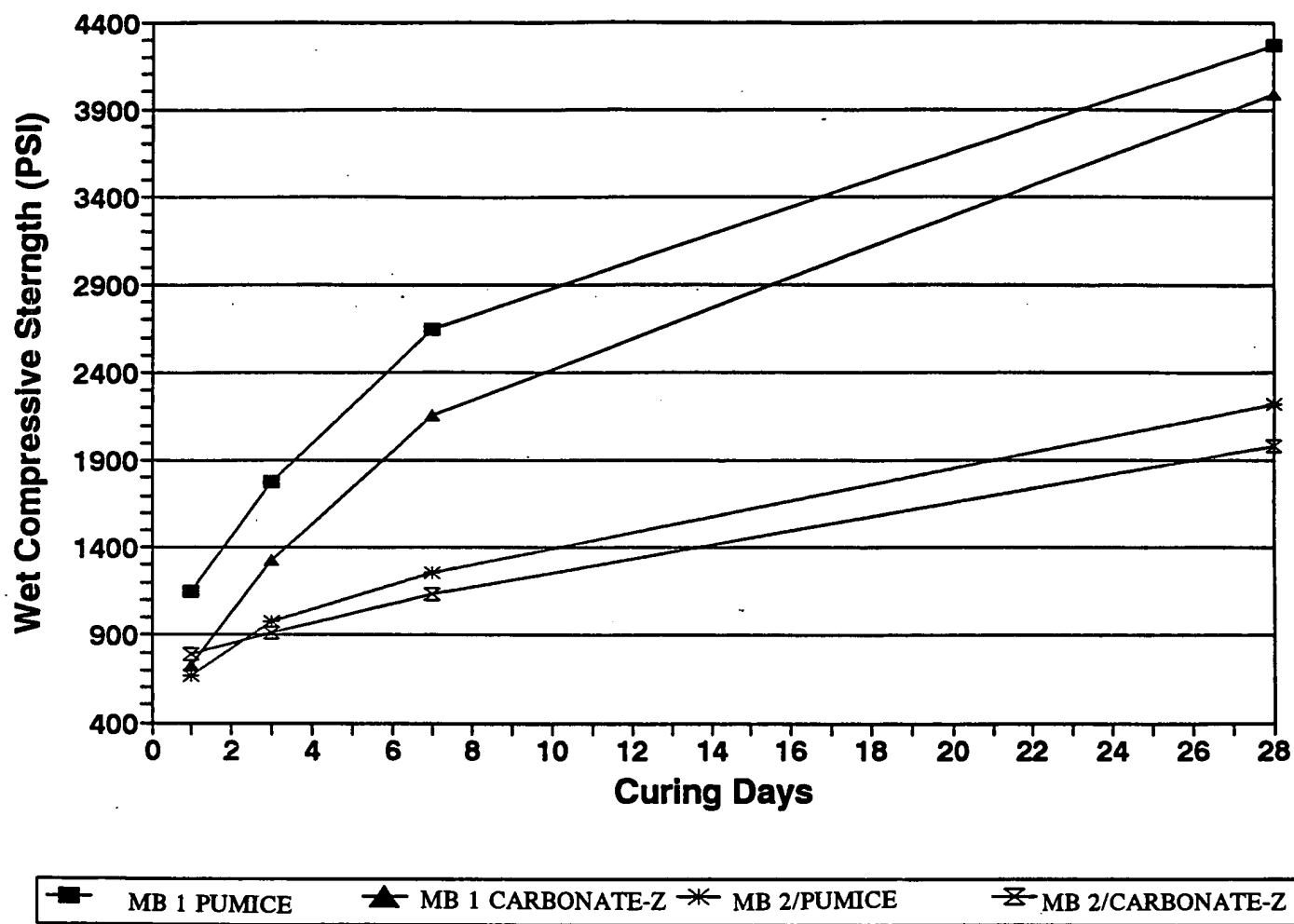
# **DECLARATION**

18. I hereby declare that all statements made herein are of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

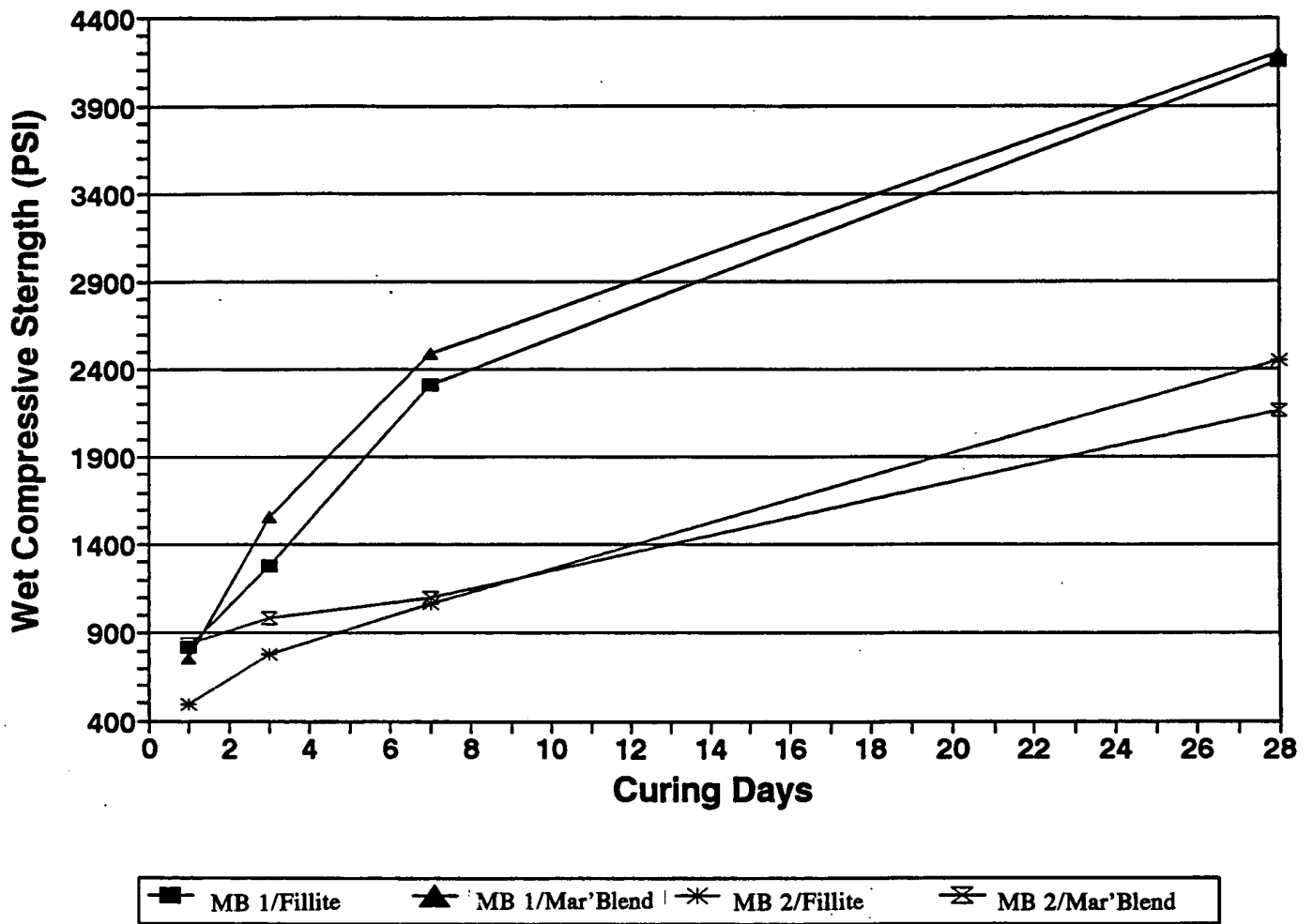
BY: Elisha Stav  
Elisha Stav

DATE: 2/7/96

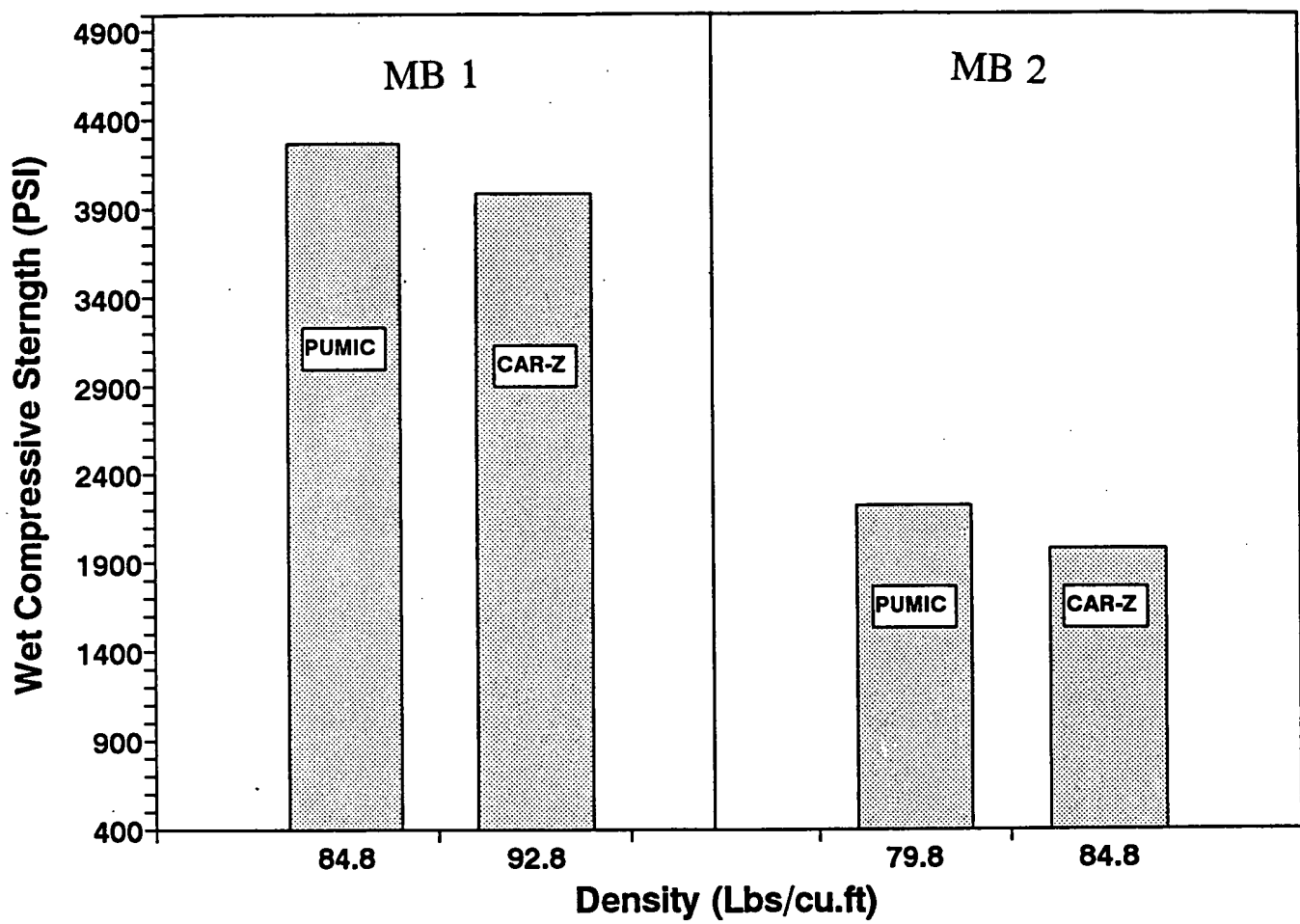
# Fig. I



**Fig. II**

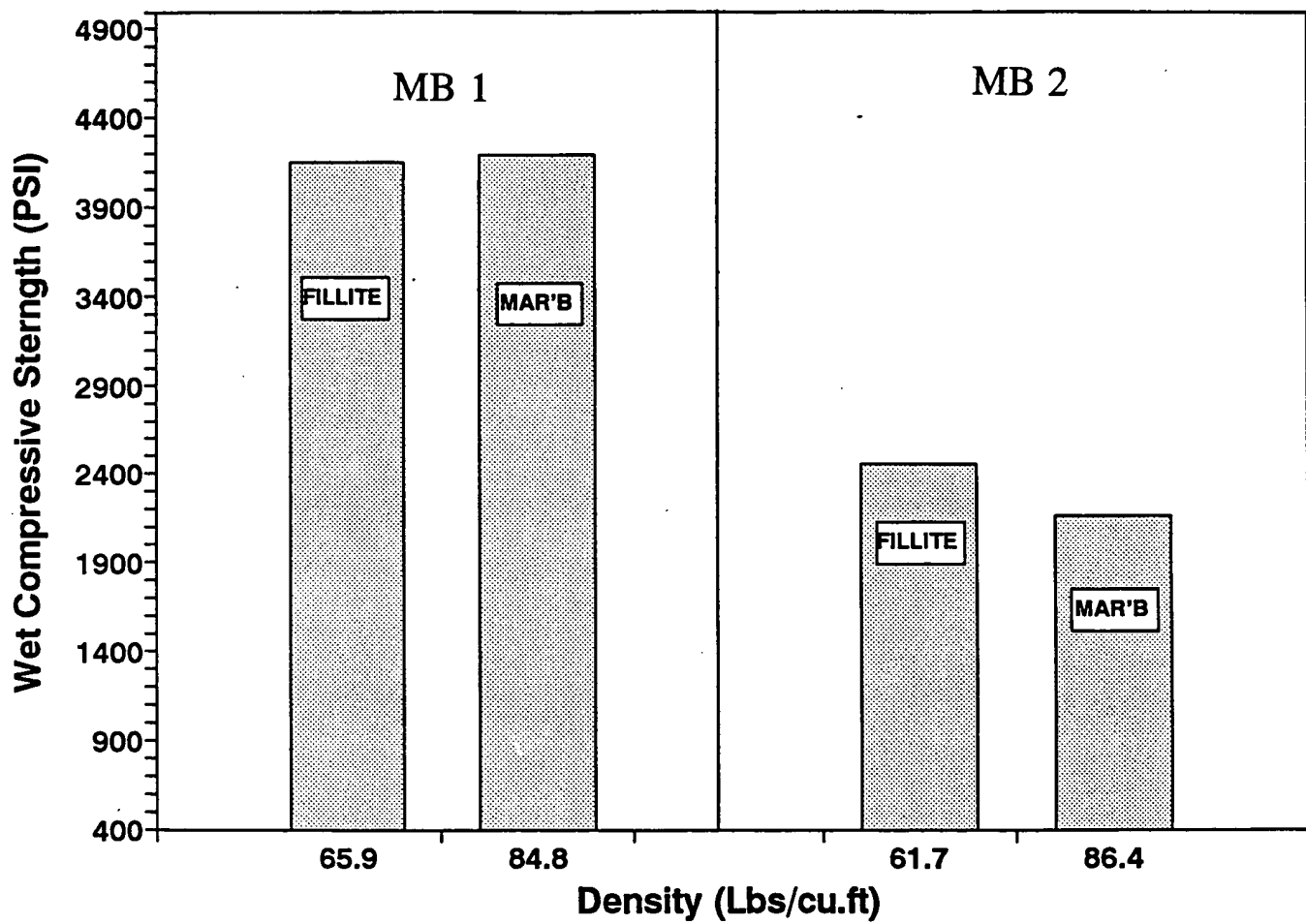


# Fig. III



(CURING 28 DAYS)

# Fig. IV



(CURING 28 DAYS)

# Fig. V

SGCSF2 ■ FILLITE PARTICLE

AUS/ON

SGCSF2

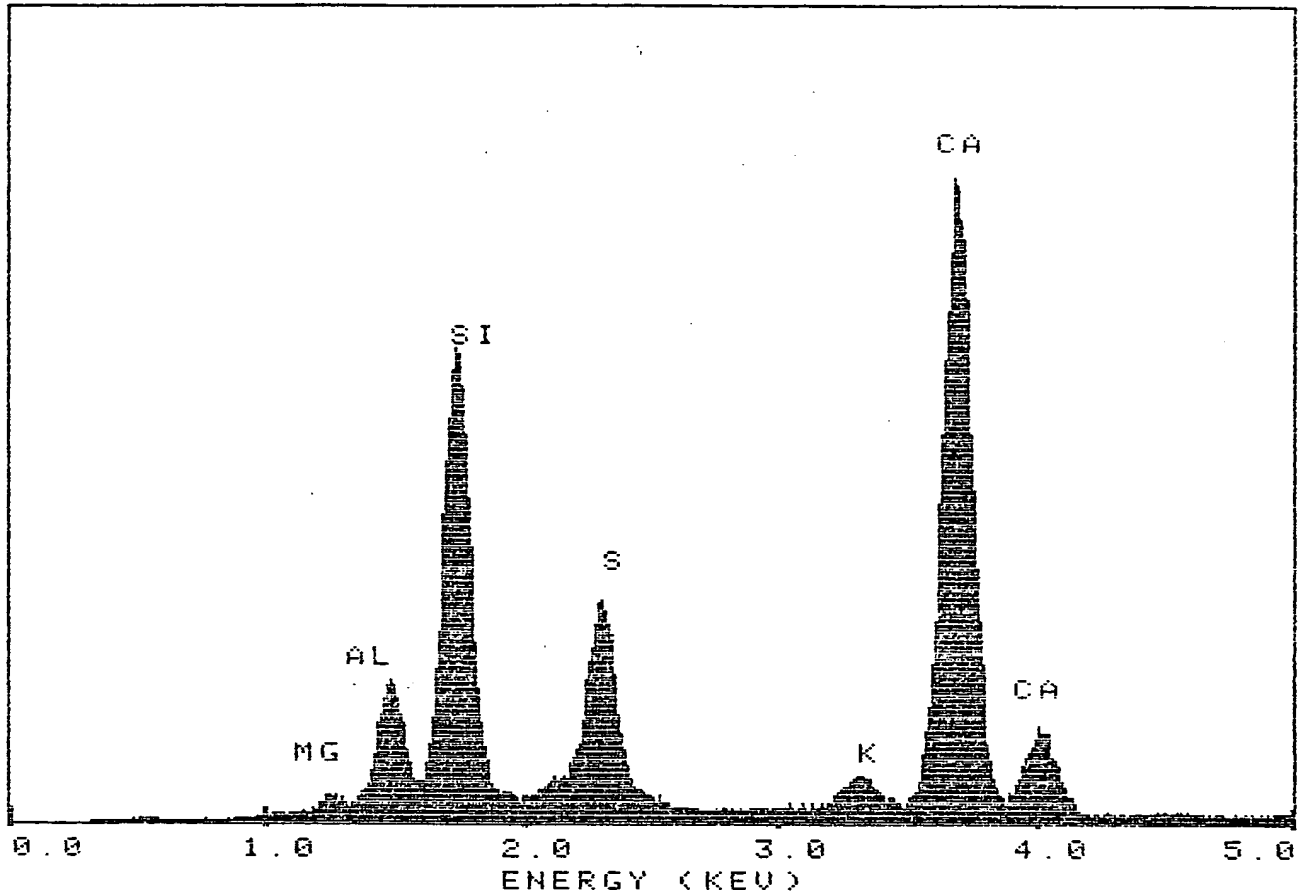
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NOSTD: STANDARDLESS ANALYSIS OF BULK SAMPLE  
SPECTRUM: GCSF2 FILLITE PARTICLE

ELEMENT	RELATIVE K	WT %
CA	0.5499	61.02
MG	0.0000	0.00
AL	0.0000	0.00
SI	0.2459	30.62
S	0.0607	8.35
K	0.0000	0.00
TOTAL		100.00

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# Fig. VI

SGCSF3 = FILLITE/MATRIX INTER

AUS/ON

SGCSF3

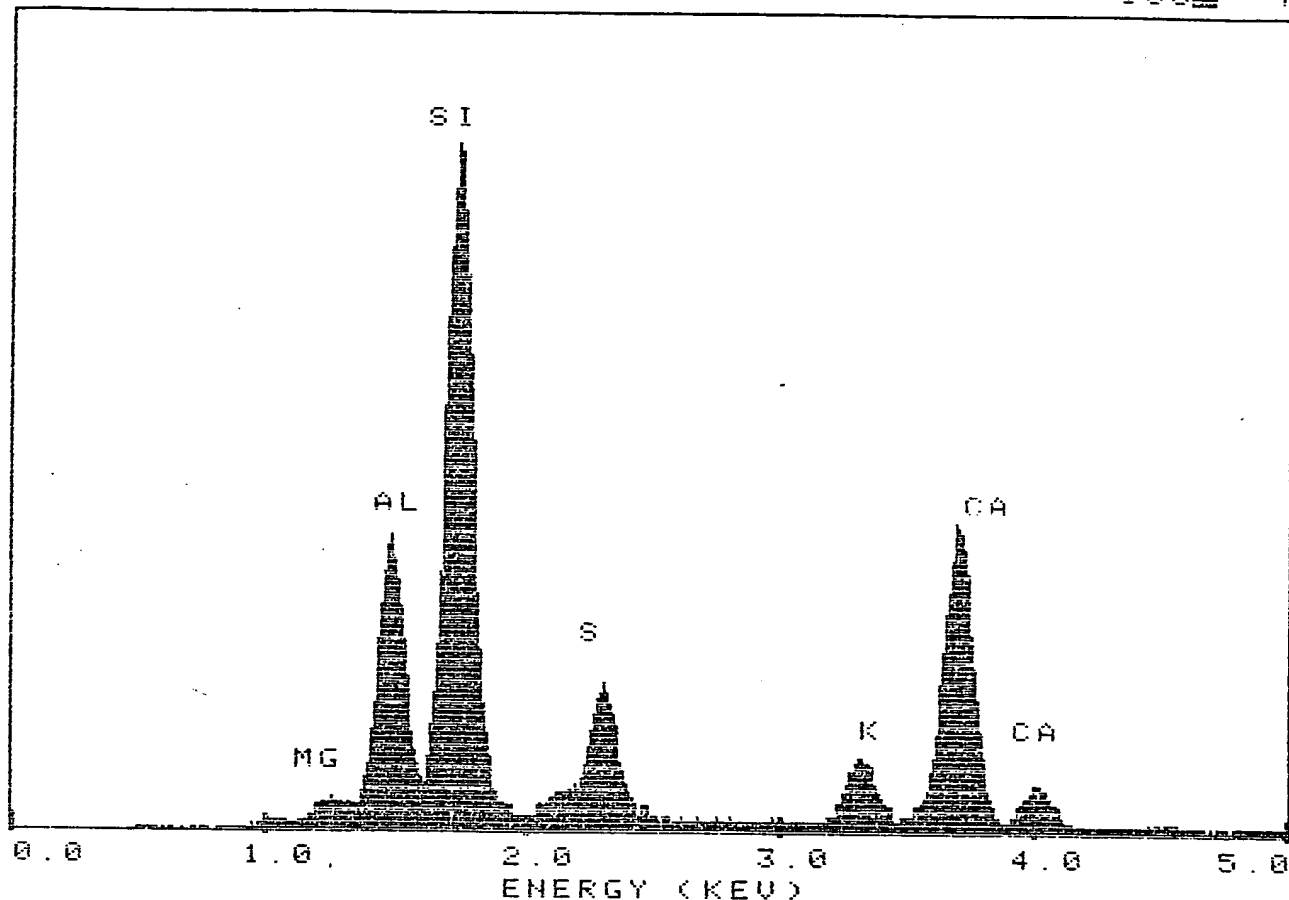
CUR:

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NOSTD: STANDARDLESS ANALYSIS OF BULK SAMPLE

SPECTRUM: GCSF3 FILLITE/MATRIX INTER

ELEMENT	RELATIVE K	WT %
CA	0.1739	21.01
MG	0.0000	0.00
AL	0.1317	15.45
SI	0.3884	53.67
S	0.0436	7.65
K	0.0175	2.22
TOTAL		100.00

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# Fig. VII

SGCSF4 ■ MATRIX NEXT TO FILLITE

AUS/ON

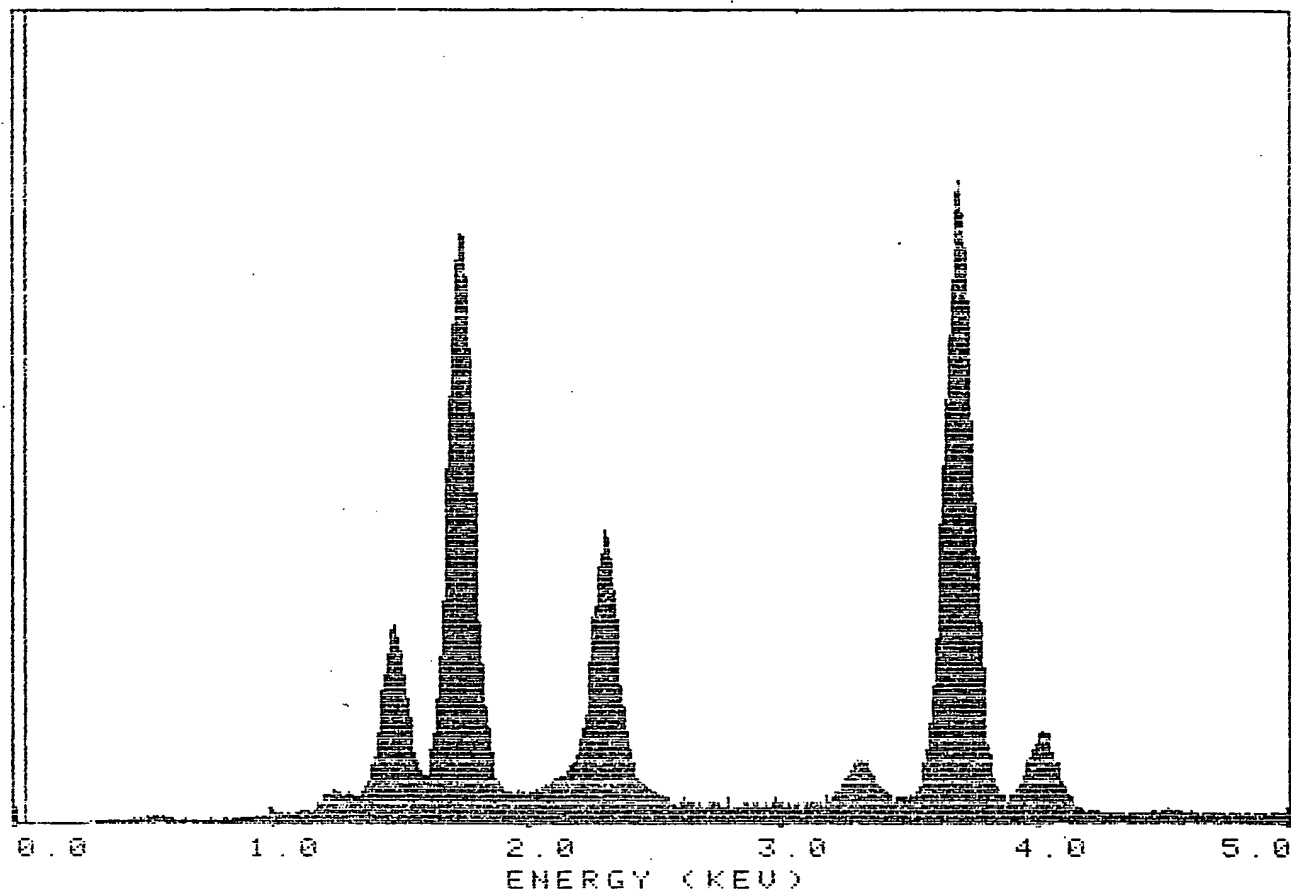
SGCSF4

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NSTD: STANDARDLESS ANALYSIS OF BULK SAMPLE  
SPECTRUM: GCSF4 MATRIX NEXT TO FILLITE  
ELEMENT RELATIVE K

CA	0.4541	WT %
MG	0.0000	0.00
AL	0.0000	0.00
SI	0.2932	35.47
S	0.0899	12.78
K	0.0000	0.00

TOTAL 100.00

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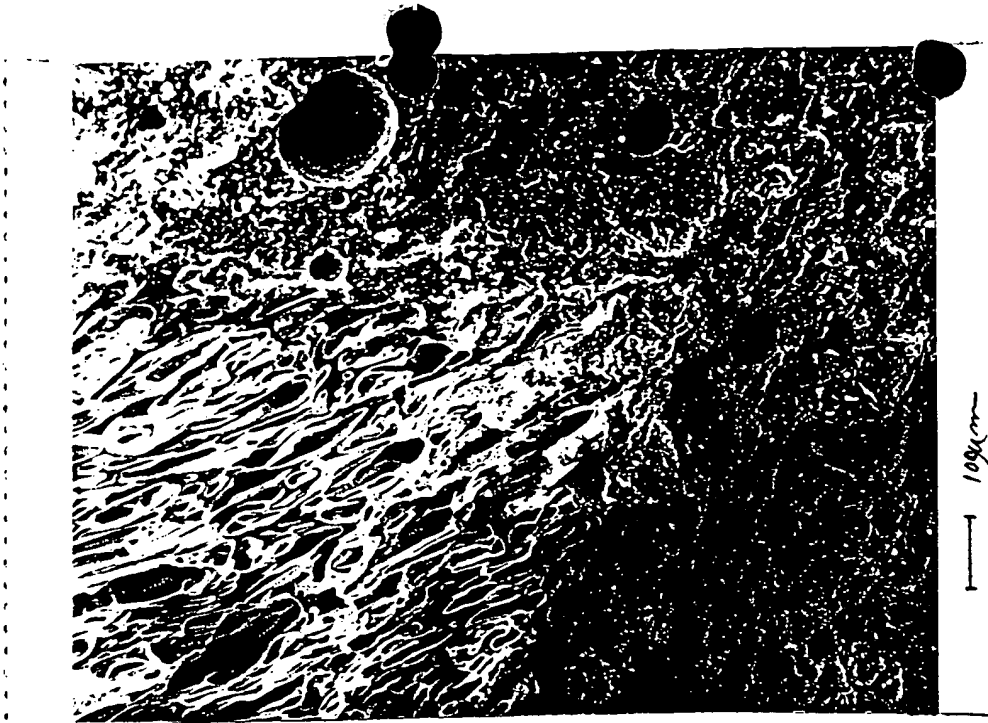


Photo 1

100x  
MAGNIFICATION

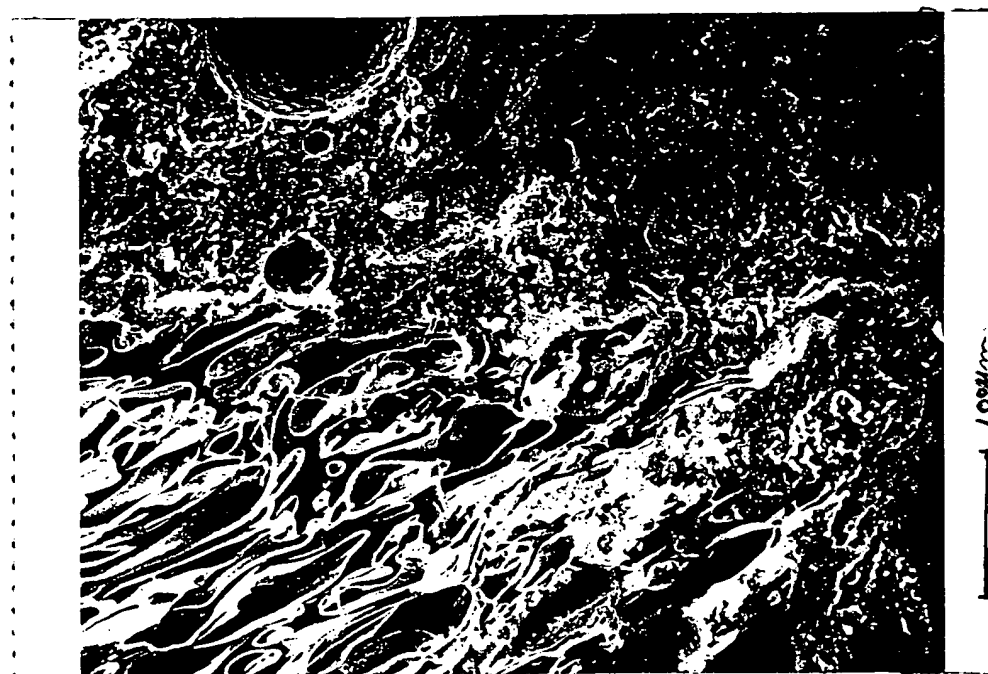


Photo 2

200x  
MAGNIFICATION

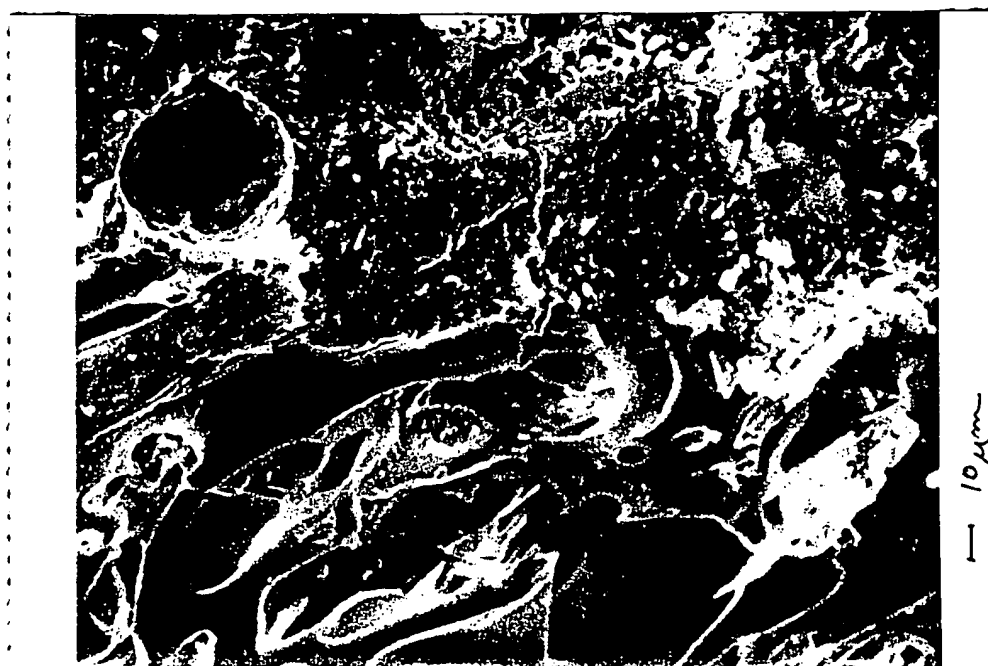


Photo 3

500x  
MAGNIFICATION

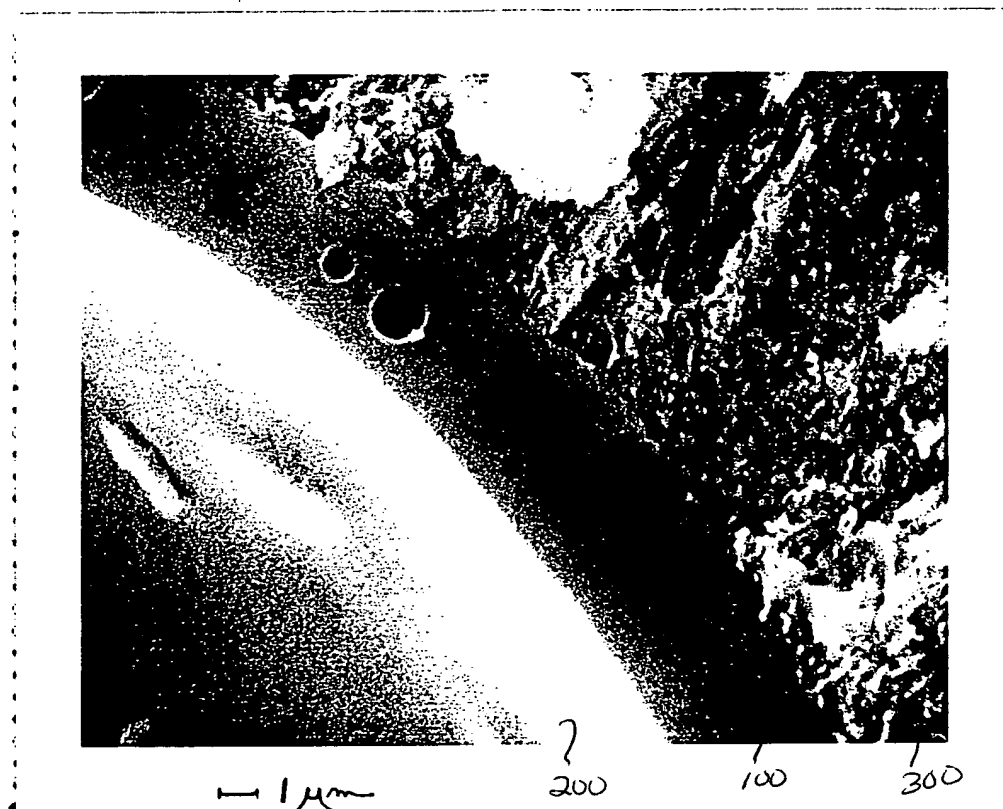


Photo 4

5,000x  
Magnification

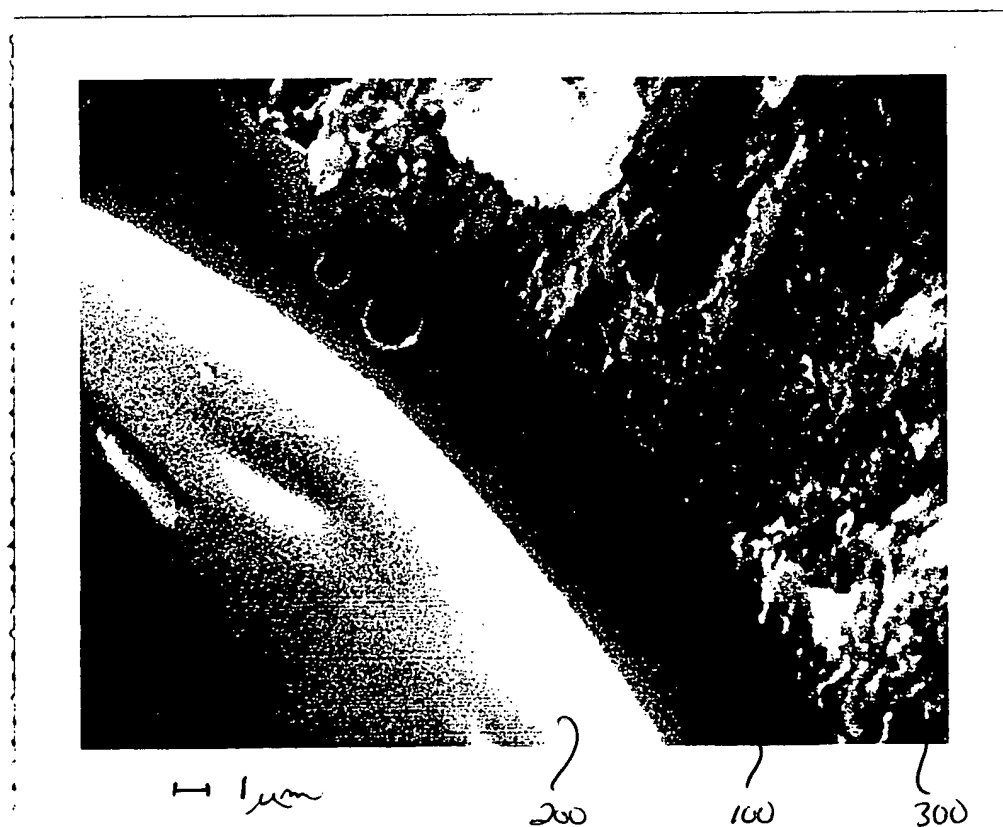


Photo 5

5,000x  
Magnification

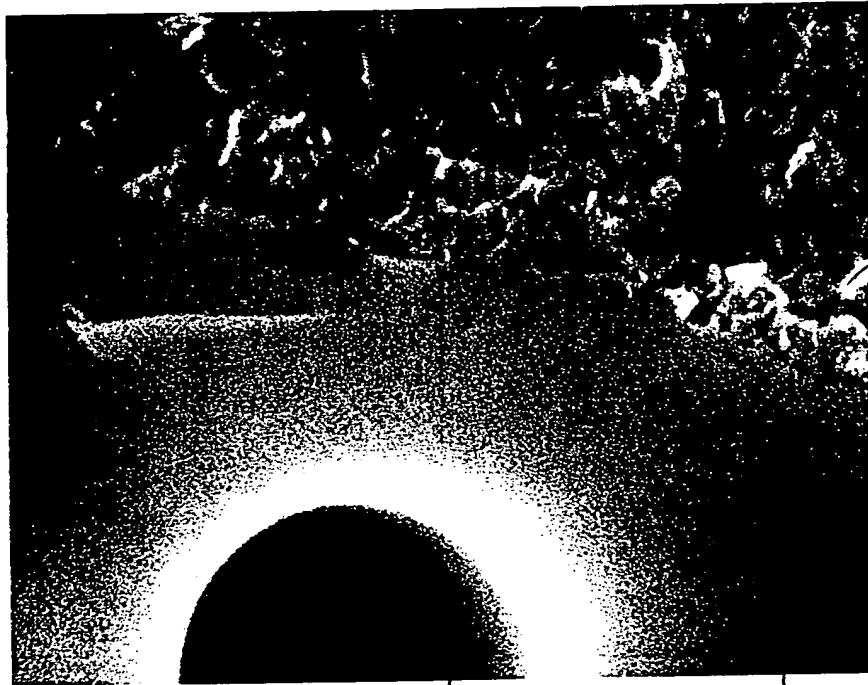


Photo 6

15,000x  
Magnification

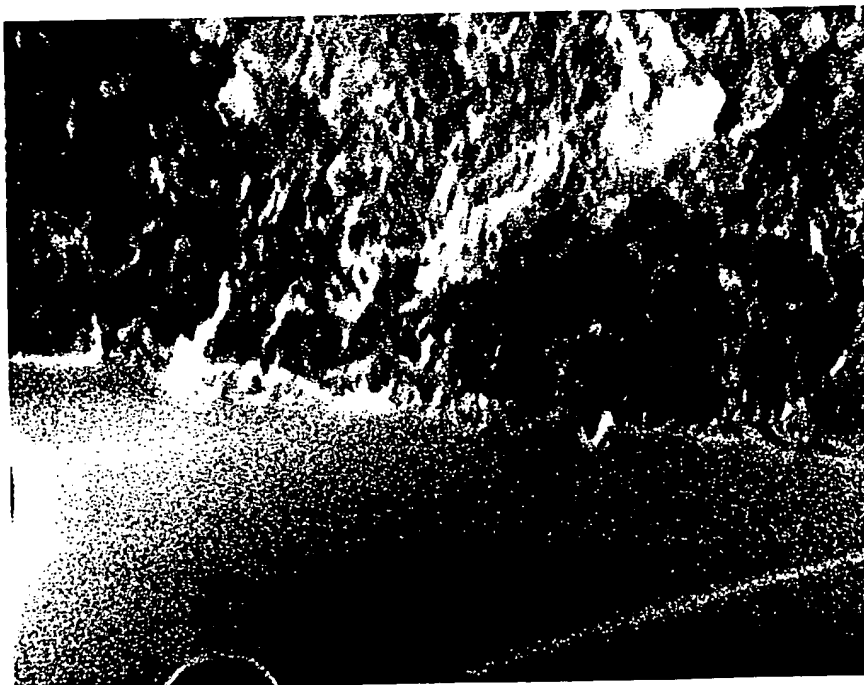


Photo 7

15,000x  
Magnification

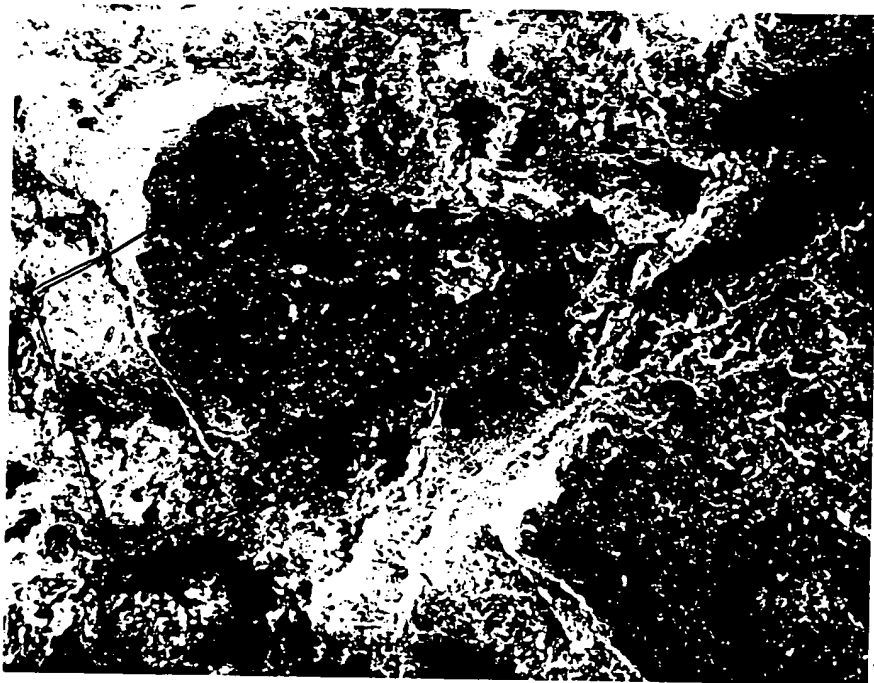


Photo 8

500x  
MAGNIFICATION